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13. ABSTRACT *(Maximum 200 words)*

The work during this contract period focused on phenomena at nanostructured metal and semiconductor surfaces, which are probed using short-pulse photoemission. Specifically, the program was to investigate the low-dimensional quantum confinement of electrons on spatially-patterned surfaces of single-crystal metals. It also aimed to expand the applications, the technique, and the instrumentation for nonlinear optical techniques of surface electrons. Electronic systems of reduced dimensionality are of interest for a variety of applications for electronic devices. In addition, the thin-film metallic structures that are currently under discussion for magnetic-memory applications, also utilize quantum confinement. This investigation has focused on the use of excited-state non-linear spectroscopy as the technique for the probing of electrons on these metal surfaces. In addition, we briefly describe a new technique probing the surfaces of semiconductors at ~ 100Å-length scale under ambient conditions.

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THE INTERACTION OF ULTRAVIOLET LASER RADIATION WITH METAL AND SEMICONDUCTOR SURFACES

FINAL TECHNICAL REPORT

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August 1, 1995 - February 28, 1998

U. S. Army Research Office

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CONTENTS

A. Statement of the Problem Studied

The work during this contract period focused on phenomena at nanostructured metal and semiconductor surfaces, which are probed using short-pulse photoemission. Specifically, the program was to investigate the low-dimensional quantum confinement of electrons on spatially-patterned surfaces of single-crystal metals. It also aimed to expand the applications, the technique, and the instrumentation for nonlinear optical techniques of surface electrons.

Electronic systems of reduced dimensionality are of interest for a variety of applications for electronic devices. In addition, the thin-film metallic structures that are currently under discussion for magnetic-memory applications, also utilize quantum confinement. This investigation has focused on the use of excited-state non-linear spectroscopy as the technique for the probing of electrons on these metal surfaces. In addition, we briefly describe a new technique probing the surfaces of semiconductors at ~ 100Å-length scale under ambient conditions. As a result, the work has built on the considerable experimental base at Columbia for the study of laser interactions at surfaces, developed on previously supported ARO Research Programs and leveraged by funding from other sponsors including NSF, AFOSR, JSEP, DOE, and AT&T.

B. Summary of Most Important Results

In this report we will first describe the development of new instrumentation for femtosecond surface probing. We will then summarize the results obtained in using this instrumentation for probing ultrasmall nanostructures on surfaces. Finally, we will provide a brief summary of recent experiments on probing semiconductor surfaces.

1. New Instrumentation for Femtosecond Probing of Surface Electron Dynamics

a) Compact and Efficient Tripled Femtosecond Ti-Sapphire Laser System

In our laboratories we have developed a new instrument for dosing time and angle resolved measurements of electron dynamics at surfaces using a femtosecond mode-locked Ti:Sapphire laser system as its backbone. Accurate surface electron dynamics studies require ultrafast-pulse time-resolved measurements with pulse widths of a few tens of femtoseconds. In addition, techniques with low laser pulse repetition rates are limited for surface dynamic measurements due to its poor signal-to-noise ratio. Ultrashort pulses at high repetition rates give

high s/n ratios even for multiphoton electron excitation in a pump-probe configuration, thus providing an excellent tool for the electronic structure and dynamics studies.

An Ar^+ laser system is used as a pump for the fs Ti:Sapphire laser system and operated at a pump power level of about 6W. As the mode-locked laser requires a spatially stable pump beam, we have developed a laser beam pointer stabilizing system[9] which corrects the temporal-spatial drift of the Ar^+ laser pump beam and maintains its spatial drift to within a few micrometers at the focal point inside the Ti:Sapphire crystal. The Ti:Sapphire laser system is a tunable source (750nm-980nm) capable of providing mode-locked laser pulses in the range of 60-120fs at a repetition rate of 90MHz and average powers ranging from 0.6W to 1W, although higher output power levels usually have a degraded spatial profile. A typical operation involves a train of 90MHz pulses of 90fs duration and average power of 0.7W (at 5.5W Ar^+ pump power) at 800nm (peak spectral intensity) corresponding to the pump conversion efficiency of η_{pump} = 13%.

The setup (see Fig. 1) also includes diagnostics which allow for an easy calibration of the Ti:Sapphire laser system as well as the second harmonic (SH) and the third harmonic (TH) beam systems; see below. These include a real-time fs autocorrelator and calibrated grating system for convenient spectrum profile measurements.

The Ti:Sapphire output laser beam is efficiently doubled (peak spectral intensity at 400nm) in a 1.5mm thick lithium triborate (LBO, LiB_3O_5) nonlinear crystal using Type I collinear phase matching (*ooe*) which allows for the non-critical phase-matching (NCPM) and thus higher conversion rates. Efficiencies as high as η_{SHG} = 33% have been obtained. The measured average output power is about 160mW at 0.6W fundamental pump, which is in excellent agreement with the theoretically predicted value, calculated from the expressions for the complex envelope of the SH radiation[10] and the SHG efficiency η

$$A_2(t,L) = -j\sigma_2 \int_0^L A_1^2\left(t - \frac{L}{v_{g2}} + \left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)x, 0\right) \cdot e^{-j\Delta kx}\,dx$$

$$\eta_{SHG} = P_2 / P_1$$

where $A_1(t,z)$ and $A_2(t,z)$ are the fundamental and the second harmonic complex envelopes, Δk is the phase mismatch, L is the crystal length, σ_2 is the nonlinear parameter given by $\sigma_2 = 8\pi^2 \cdot d_{eff}/(n\lambda)$, where d_{eff} is the effective nonlinear coefficient and λ is the wavelength of the

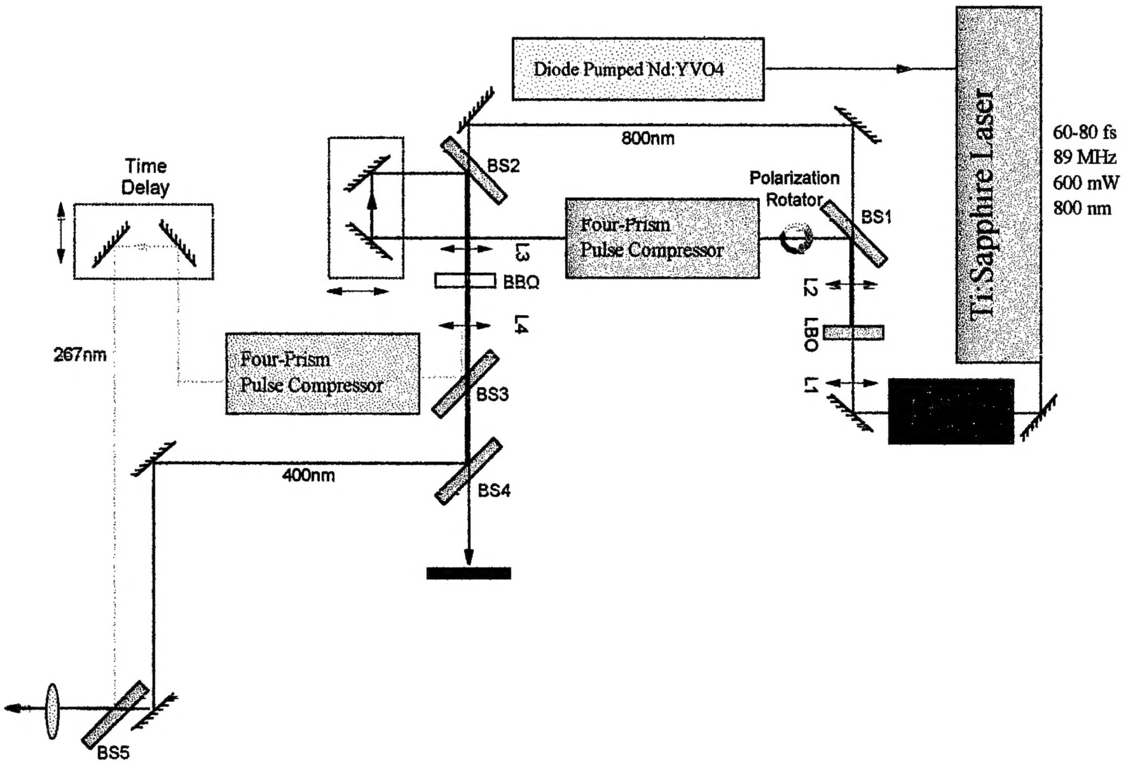


Figure 1. Femtosecond optical apparatus for probing electronic dynamics near nanostructures.

fundamental beam. v_{g1} and v_{g2} are the group velocities, and P_1 and P_2 are the average power levels at ω_1 and ω_2, respectively. In the calculation, the *sech* profile has been assumed for the complex envelope at the fundamental wavelength.

The generated SH and the fundamental beam are further frequency-mixed in a second 0.5mm thin nonlinear crystal, beta barium borate (BBO, (β-BeB_2O_4), using Type-I phase matching (*ooe*). The output signal, the third harmonic (peak spectral intensity at 267nm), has an average power of 40mW corresponding to the efficiency of $\eta_{THG} = 27\%$ with respect to the SH

signal level. We are currently testing a much thinner BBO crystal so as to achieve resolution at even high short time scales.

As they propagate in dispersive media (focusing and reflecting elements in the setup, LBO and BBO crystals), all three wavelength beams broaden, thus reducing their peak power levels and increasing pulse widths (to a few hundreds of fs). We have therefore setup a four-prism sequence pulse compressor, which dramatically restores the third harmonic pulse widths to the initial pulse width values of the fundamental beam. The SH (400nm) and the TH (267nm) pulses are then controllably time-delayed. The fundamental SH and TH pulses are focused onto the sample.

Monochromatic spectrum have been taken from a Cu(111) surface with the TH UV pulses to give excellent s/n ratio for the n=1 image state. The photon energy was 4.6seV and the average power was 25mW. The spectrum was taken with a 127° spherical-sector energy analyzer. The setup also enables us to do angle-resolved measurements. Further time-resolved experiments on Cu have shown controllable, fs-scale time pump-probe measurements can be made.

b) Instrumentation for Surface Analysis and Angle- and Time-Resolved Nonlinear Photoelectron Spectroscopy

Our instrumentation for the measurements of the excited electronic structure at surfaces and interfaces has been improved significantly over the last two years it now represents a unique university instrument for these measurements. The system now consists of: i) an enhanced UHV (ultrahigh vacuum) chamber suitable for multitechnique measurements, ii) basic surface cleaning and diagnostic tools: an ion sputter gun, LEED (low energy electron diffraction) for lattice structure analysis, and AES (Auger electron spectroscopy) for chemical composition at the surface, iii) a UHV compatible evaporator, which allows *in situ* growth of various layered structures with thicknesses of from submonolayer to thousands-of-Å's, iv) an UPS (ultraviolet photoemission spectroscopy) system for measurement of initial, occupied electronic states, providing information needed to assess the occupied electronic structure at the surface/interface, and v) two electron-energy analyzers. Regarding the latter, the two systems consist of a compact, high-resolution analyzer, suitable for angle-resolved measurements in a wide range of angles, and a larger, 100mm hemispherical analyzer; useful in high-sensitivity UPS, AES as well as in high-resolution nonlinear photoemission measurements.

2. Measurements of the Excited Electronic Structure of Low-Dimensional Angstrom-Size Features

a) Coherent Electron Confinement by a 10Å Lateral Superlattice

Low-dimensional surface systems have attracted much attention recently because of their fundamental and technological implications. Traditionally, work in these systems has utilized either two-dimensional confinement,[1] that available at heterojunctions in electronic devices,[2] or for lower dimensionality, through lithographic patterning of 'wires' or 'dots' on single-crystal surfaces.[3] Metallic systems with spacer-layer structures have been observed to possess quantum-well states[4] and display effects of oscillatory magnetic coupling[5] as seen in superlattices potential applications in magnetic storage.[6] Low dimensionality can be effectively realized and utilized[7-11] via the use of the natural atomic-scale features on vicinally cut surfaces, such as the stepped surfaces created by a small-angle miscut from a low-index plane on a single-crystal metal. The extremely small length scales in this case means that high confinement energies can be obtained, and hence operation at room temperature is possible.

Recently, using our nonlinear photoemission system, we have made the first direct observation of the formation of a lateral superlattice, effects the movement of electrons parallel and just above the stepped surface. Our results show that the lateral periodicity (~11Å) of the step potentials on a bare stepped surface leads to back-folding of the electrons dispersion within the surface Brillouin zone. A surface preparation, which pins the surface steps with impurity atoms, sharpens this characteristic dispersive behavior to the point that it can be followed to the edges of the lateral Brillouin zone formed by the step lattice. The resulting reduction of the surface Brillouin zone by a factor of 4.5, compared to that of planar Cu(001), causes an oscillatory dispersive behavior where the electron energy is a multivalued function of k as seen in the first two Brillouin zones of such a lateral superlattice formed by the regular steps. One-dimensional superlattice effects appear to have not been seen previously in the band structure of nanostructured surfaces. Our results are remarkable in that the quantum confinement seen in this case is at dimensions much smaller than those typically seen in the usual electronics materials system.

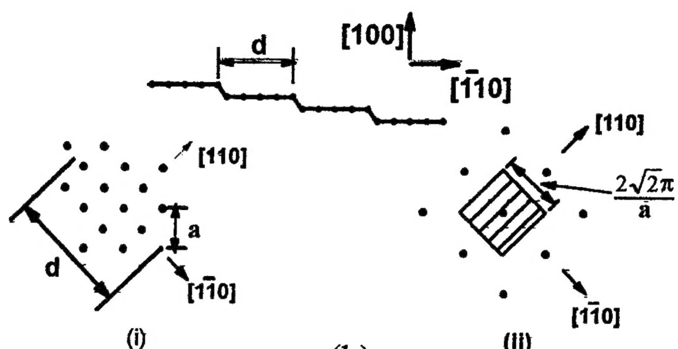
The dispersive behavior measured in our case is shown in Fig. 2(a) and is in accord with that expected from a lateral surface superlattice, which can be provided by a stepped metal

surface with a periodic modulation of the surface potential from the steps. A sketch of the relevant real space lattice and Brillouin zones on the stepped Cu(001) is shown in the inset of Fig. 2(b). For image-state electrons, the effective magnitude of the step potential depends on the average distance of the electron from the crystal plane; in fact, it is interesting that step-edge perturbation in the surface potential is sufficient to form an electronic structure of a lateral superlattice. Electrons in such a 1D periodic potential exhibit new Brillouin zones, whose extent in k-space is determined by the reciprocal step-lattice vector of $g=2\ \pi/d$. The electrons excited to the image state will experience Bragg reflections, which result in a multivalued dispersion function which repeats itself at every multiple of $k = \pi/d$, *within* the *first* surface Brillouin zone (0 to 1.23 $Å^{-1}$) of the flat (001), as seen in Fig. 2(a). In addition, 1D Kronig-Penny calculations show that the superlattice potential would also be expected to provide lateral confinement for the image-state electron leading to a larger effective mass.[12]

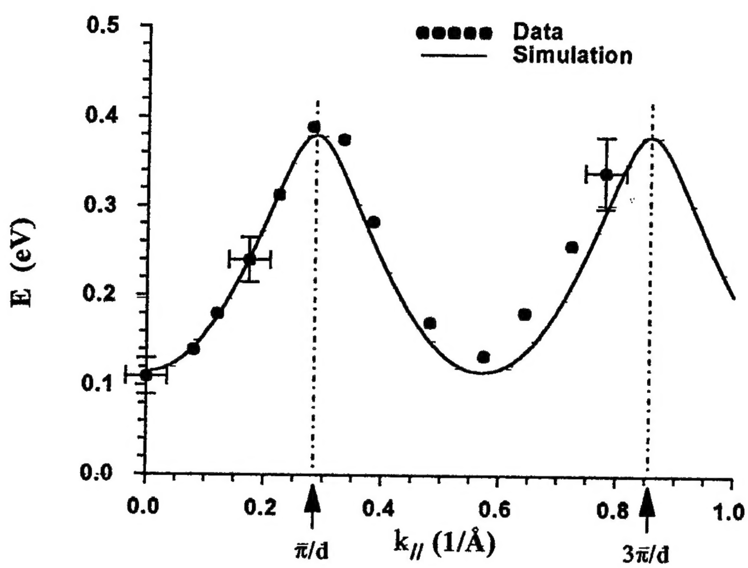
b) How Electrons Move Near Nanostructures

Electronic confinement and movement is central to a variety of novel electron and magnetic nanoscale devices. There has been a recent growth in research interest in electron motion and electronic structure in the vicinity of nanoscale features on single-crystal metals.[8,9,13] Lateral quantum confinement of electrons on metal surfaces was seen resulting in sharp images of electron standing waves in the vicinity of surface scattering centers, such as Angstrom-high step edges, through STM studies.[9] In conjunction with the interest in lateral confinement of *sp* surface-state electrons, several recent questions have arisen on the magnitude and nature of scattering at the lateral barrier.[14]

In our investigation of electron confinement by nanoscale features at or near the surface, we have used measurements of the dispersion of a well characterized series of surface states to characterize the response of surface electrons to a regular array of steps on a single-crystal copper surface, Cu(775). Since each of the states has different average distances above the crystal surface, the measurements provide insight into the effects of step potential at three values of z. We probed the n=0 (occupied) and the n=1,2 (unoccupied) states, in the terminology of Smith,[15] on both flat Cu(111) and stepped Cu(775) for comparative studies. The n=0, crystal-induced surface state is located within the plane of the last lattice points while the n=1 and n=2 surface Rydberg states (or image states) have an average location at 3 and 12 Å respectively



(b)



(a)

Figure 2(a). The dispersion relation of the lateral superlattice formed on N-decorated, stepped Cu(001). The filled circles are our data, the full line is a fit, and the dotted lines indicate the Brillouin zone boundaries of the step superlattice. The back-folded band between π/d and 3 π/d is absent on the flat Cu(001) surface; (b) a sketch of the i) real space lattice and ii) Brillouin zones on the stepped Cu(001), where the first Brillouin zone of the planar Cu(001) (bound region) contains 4.5 Brillouin zones of the lateral superlattice in the [110] direction.

from the jellium edge. Our measurements of both the occupied and unoccupied states have used angle-resolved *resonant* monochromatic and bichromatic two-photon photoemission (2PPE). By tuning the photon energy and photoelectron detection angle, resonant excitation from the n=0 state to n=1,2 states could be achieved and hence the energy bands of both the initial and intermediate states could be mapped out.

Figure 3 shows the energy dispersion curves (as a function of electron momentum parallel to the surface) of the n=0,1, and 2 states on stepped Cu(775), derived from numerous resonant 2PPE measurements.[16] Note that on flat Cu(111), all the three dispersion curves are symmetric about $k_{\parallel} = 0$ or the surface normal (not shown here). In contrast, Fig. 3 shows that on stepped Cu(775) the dispersion minimum of both the n=0 and n=1 states are shifted in the k-space. However, in the case of the n=2 state the dispersion curve was still centered at $k_{\parallel} = 0$. The energy band minimum of the n=0 state was found to be located at $k_{\parallel} \sim 0.22$ Å^{-1}, corresponding to a + 17° detection angle for photon energies close to 2.18 eV; and the n=1 state minimum was shifted to $k_{\parallel} \sim 0.09$ Å^{-1}, or +8~9°, a value which corresponds to the direction of the terrace normal, (111). Such results show that electrons at different heights, even within a nanometer, from the surface exhibit different dispersive behaviors, which is in accord with the nature of the interactions with the surface steps. Specifically, the n=1 electron is oriented to the (111) terrace while the n=2 state, located farther away from the surface, is oriented to the general (775) surface. The embedded n=0 state dispersion was found to be determined by the bulk band projection onto the specific (775) surface and not related to the magnitude of the step potential.

3. Two-Photon Photoemission from Si(111)

Physical understanding of the transient phenomena of semiconductors is very important for developing future-generation microelectronic technology. Fundamental studies of hot carrier dynamics are essential to predict the performance and reliability of the ultrafast semiconductor devices. Many surface chemistry phenomena such as absorption and desorption are also related to the transient processes in semiconductors. As a result, studies of electron dynamics in crystals have attracted much attention for the last decade and have included studies on metals, semiconductors with direct or indirect band gap, as well as reconstructed surfaces such as Si(100)2x1, Si(111)2x1, etc.

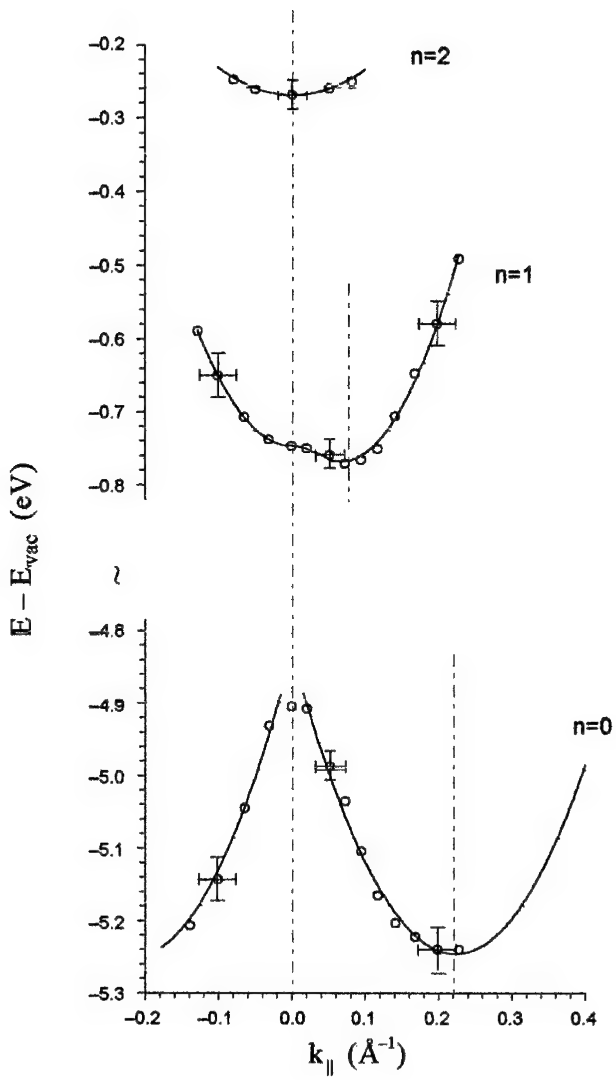


Figure 3. Energy dispersion curves of the n = 0, n = 1, and n = 2 states on stepped Cu(775).

Time-resolved multiphoton photoemission spectroscopy has proven to be a particularly powerful technique for probing carrier dynamics at surfaces and interfaces. We have probed the electron dynamics at the Si(111)7x7 surface using angle- and time-resolved two-photon photoemission experiments. In these experiments, either a monochromatic or a bichromatic pump-probe beam configuration was used. The pump laser pulses populate the conduction states and unoccupied surface states, the lifetime of which is in the range of tens of femtoseconds. The photoelectrons on these states are then excited by the probe pulse, with no time delay. We are able to study the decaying channels of hot electrons if a time delay is preset between the probed and pump pulses. Nanosecond pulses with tunable photon energy were first used to study the unoccupied electronic states on Si(111)7x7 surface. By varying the polarization of the probe light, we observed a distinct difference of the photoemission signals with respect to the type of polarization (s or p). The results provided a strong experimental support that the "CBM shoulder," reported earlier by M.W. Rowe, et al., does in fact come from a surface resonance. This relatively long lifetime unoccupied surface state has also been examined by inverse photoemission.

The electron dynamics can be further studied by femtosecond laser pulses. The experimental setup used a variable time delay between femtosecond (fs) pump and probe pulses, which were generated by frequency doubling and tripling laser pulses from a Ti:sapphire fs laser system. This setup which enables the relaxation channels of electrons excited above the conduction-band minimum (CBM) to be measured with subpicosecond time resolution is currently being tested.

4. Recent Experiments

Since the end of this contract we have completed several experiments using our Femtosecond pump-probe experiments. The results in these experiments, two examples of which are shown in Figures 4 and 5, are briefly summarized in our recent talks for the Centennial APS meeting. We include the abstracts for these talks in the Appendix.

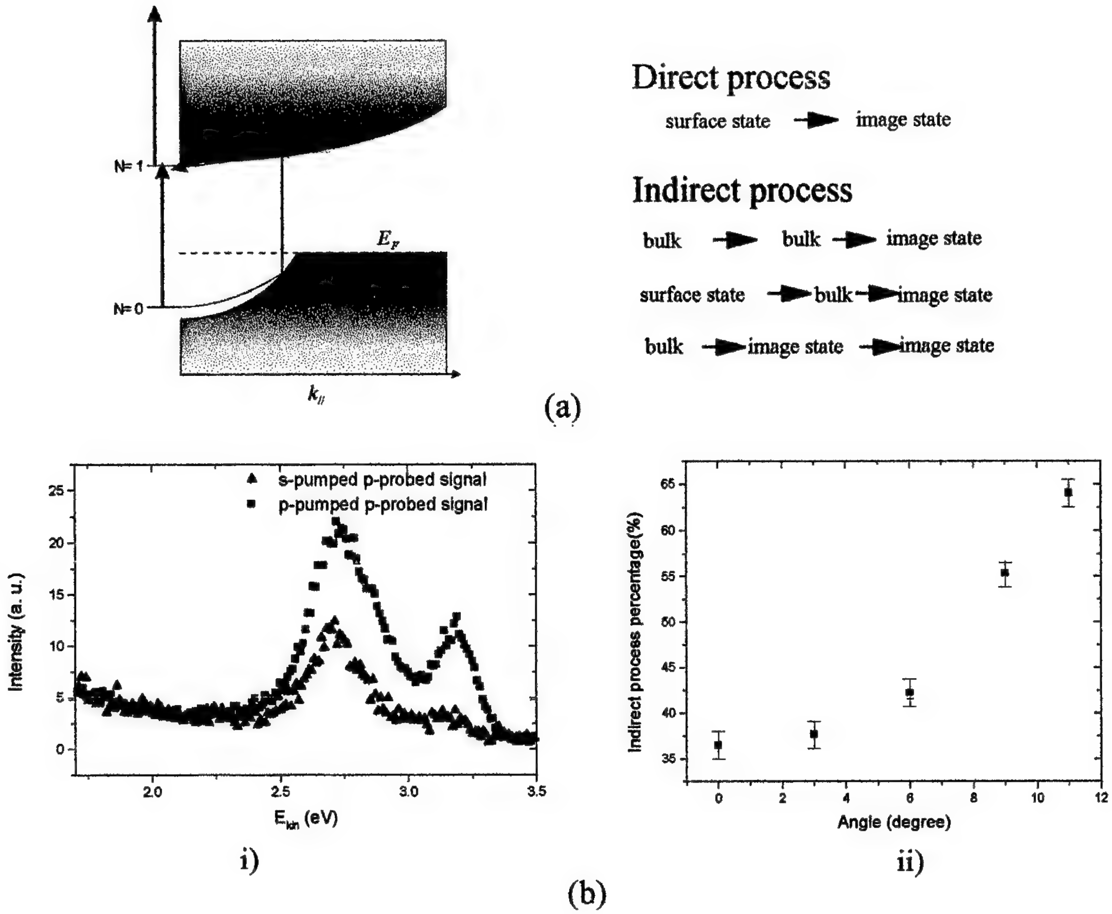


Figure 4. (a) Excitation mechanisms in two-photon photoemission on Cu(111). (b) i) Comparison between p-pumped and s-pumped signal on Cu(111). S-pumped signal comes from indirect process and p-pumped signal comes from both direct and indirect processes. ii) Percentage of indirect transition at different angles on Cu(111).

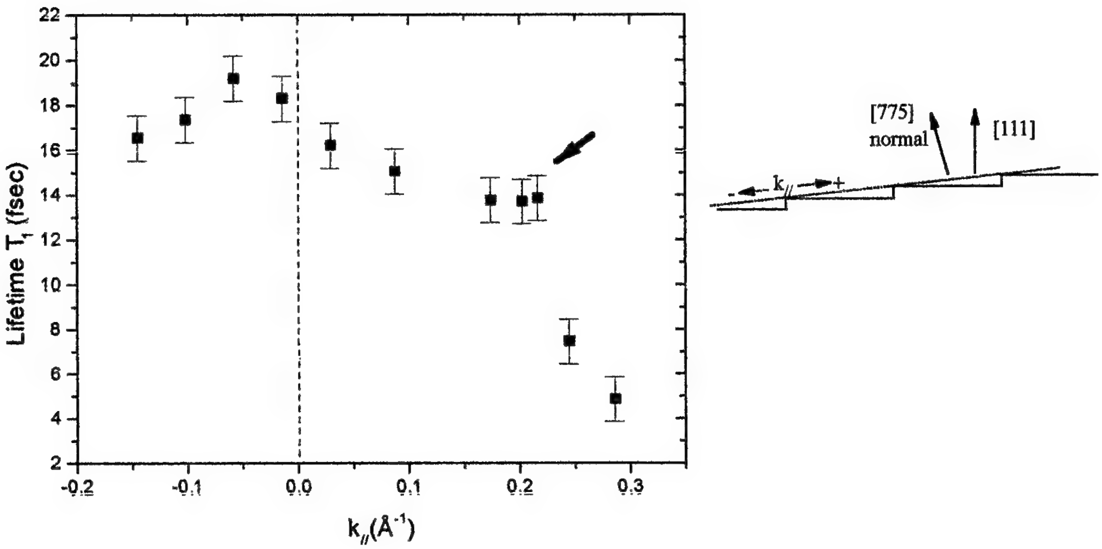


Figure 5. Momentum-resolved lifetimes of n=1 image state on stepped Cu(775).

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6. List of All Participating Scientific Personnel (and those showing advanced degrees)

Charlie Wang - Postdoctoral Research Scientist
H. Kwak - Postdoctoral Research Scientist
Antonije Radojevic - graduate student
Shawn Shen - graduate student
Gabor Nagy - Postdoctoral Research Scientist
Steve Laufer - undergraduate student
Kevin Aptowicz - undergraduate student

7. Publications & Presentations

1. R.M. Osgood Jr. and X. Y. Wang, Chapter in *Solid State Physics*, v. 51, "Image States on Single-Crystal Metal Surfaces," H. Ehrenreich and F. Spaepen, eds.; Academic Press, New York, (1998).

2. L.-L. Chao, G.S. Cargill III, M. Levy, R.M. Osgood Jr., and G.F. McLane, "Cathodoluminescence Study of GaAs Quantum Wells and of Submicron Dots Fabricated by Magnetron Reactive Ion Etching," Appl. Phys. Lett. 70, 408 (1997).

3. X.-Y. Wang, X.J. Shen, and R.M. Osgood Jr., "Surface Electron Motion Near Monatomic Steps: Two-Photon Photoemission Studies on Stepped Cu(111)," Phys. Rev. B. 56, 7665 (1997).

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Presentations to Industry and Symposia

1. THE AMERICAN PHYSICAL SOCIETY, (Centennial Meeting) Atlanta, GA, March 20-26, 1999
"Two-Photon Photoemission Study of Stepped Cu(111) Surface Using Femtosecond Laser Pulses," X.J. Shen, H. Kwak, A.M. Radojevic, and R.M. Osgood, Jr.

2. THE AMERICAN PHYSICAL SOCIETY, (Centennial Meeting) Atlanta, GA, March 20-26, 1999
"K-dependence of Image State Lifetime Measurements on Cu(111) Surface with Time-resolved and Angle-resolved Photoemission," H. Kwak, A.M. Radojevic, X.J. Shen, R.M. Osgood, Jr.

3. AMERICAN VACUUM SOCIETY MEETING, Baltimore Convention Center, Baltimore, MD, November 2-6, 1998
"Chemically Assisted Ion-Beam Etching of Submicrometer Features in GaSb-based Quantum Wells," G. Nagy, R.U. Ahmad, M. Levy, R.M. Osgood, Jr., M.J. Manfra, and G.W. Turner.
4. CLEO/QELS '99, Baltimore, MD, May 23-28, 1998

"Momentum-Dependent Ultrafast Dynamics of Image States on Flat and Stepped Cu(111) Surfaces," X.J. Shen, H. Kwak, A.M. Radojevic, and R.M. Osgood, Jr.

5. 1997 MARCH MEETING OF THE AMERICAN PHYSICS SOCIETY, Kansas City, MO, March 17-21, 1997
"Effects of Metal Nanostructures on Surface Electrons," X.Y. Wang, X.J. Shen, and R.M. Osgood, Jr.

6. WORKSHOP ON ULTRAFAST SURFACE DYNAMICS, Ascona, Lago Maggiore Switzerland, March 2-5, 1997
"The quantum confinement of surface electrons by metal nanostructures," X.Y. Wang, X.J. Shen, and R.M. Osgood, Jr.

7. OPTICAL SOCIETY OF AMERICA, Santa Fe, New Mexico, February 9-14, 1997
Winter '97 Topical Meetings, "Surface Electronic Structure on Nanostructured Metal Surfaces," X.Y. Wang, X.J. Shen, F. Huang and R.M. Osgood, Jr.

8. GORDON RESEARCH CONFERENCE, Henniker, New Hampshire, June 23, 1996
Nanostructure Fabrication,
"Cathodoluminescence and Photoluminescence Study of Submicron Features Fabricated by Magnetron Reactive Ion Etching," Miguel Levy (poster, not talk).

9. 56th PHYSICAL ELECTRONICS CONFERENCE, Boston University, Boston, MA, June 17-19, 1996
"Height-Dependent Surface Electronic Structure on Stepped Cu(775)," X.Y. Wang, X.J. Shen, and R.M. Osgood, Jr.

10. AMERICAN PHYSICAL SOCIETY ANNUAL MEETING, St. Louis, MO, March 19-22, 1996
"Evidence for Electron Localization by the Step-Edge Potential for Electrons in the n=1 Image State on Cu(111)," X.J. Shen, X.Y. Wang and R.M. Osgood, Jr.

11. AMERICAN PHYSICAL SOCIETY ANNUAL MEETING, St. Louis, MO, March 19-22, 1996
"One-Dimensional Coherent Effects on Vicinal Cu(001)," X.Y. Wang, X.J. Shen, R. Haight, F.J. Himpsel and R.M. Osgood, Jr.

12. AMERICAN PHYSICAL SOCIETY ANNUAL MEETING, St. Louis, MO, March 19 - 22, 1996
"A Comparison of the Energy Dispersion of the n=1 and n=2 Image States on Flat and Stepped Cu(111)," X.Y. Wang, X.J. Shen, and R.M. Osgood, Jr.

13. AT&T BELL LABS SEMINAR, Holmdel, NJ, November 21, 1995
"Image States on Single-Crystal Metals," Richard M. Osgood, Jr.

14. AMERICAN PHYSICAL SOCIETY, Flagstaff, AZ, June 12-14, 1995
55th Conference on Physical Electronics,
"Reduced-Dimensional Electron Confinement on Stepped Cu(100)," X.Y. Wang, X.J. Shen, and R.M. Osgood, Jr.

15. 1995 QUANTUM ELECTRONIC LASER SCIENCES CONFERENCE, Baltimore, MD, May 22-26, 1995
"A Two-Photon Photoemission Study of Two-Dimensional Scattering Processes on Na-Covered Cu Surfaces," X.Y. Wang, R. Paiella and R.M. Osgood, Jr.

16. M.I.T. DEPARTMENT OF MATERIALS SCIENCE & ENGINEERING, Cambridge,MA, March 16, 1995, Electronic Materials Thursday Seminar,
"Advanced Etching Technologies for Integrated Optics and Quantum-Confined Structures," Richard M. Osgood, Jr.

17. DUKE UNIVERSITY, Durham, NC, February 15, 1995
Physics Department Colloquium, "Image Electrons Near Single-Crystal Surfaces," R.M. Osgood, Jr.

APPENDIX

Abstract Submitted
for the MAR99 Meeting of
The American Physical Society

Sorting Category: 14.5 (Experimental)

K-dependence of Image State Lifetime Measurements on Cu(111) Surface with Time-resolved and Angle-resolved Photoemission HIDONG KWAK, A. M. RADOJEVIC, X. J. SHEN, R. M. OSGOOD JR., Columbia Radiation Laboratory, Columbia University — We used femtosecond time-resolved and angle-resolved two-photon photoemission spectroscopy to study dynamics of image states on Cu(111) surface. This is the first k-space state resolved study of image states lifetimes. Our experiments were conducted on a clean Cu(111) prepared in an ultrahigh vacuum chamber. The output of Ti:sapphire oscillator was frequency doubled and tripled to generate 3.1 eV and 4.65 eV beams using nonlinear crystals. An 80 fs, 400 nm and a time delayed 100 fs, 266 nm laser beams were coincident on the sample at 70 degrees angle of incidence. Photoemitted electrons due to the pump and probe correlation were detected as a function of time-delay. The peak of the correlation signal is delayed with respect to the zero time delay, which is found by measuring the signal of the direct transition via virtual intermediate states. This delay is due to the finite lifetime of the intermediate image states. Quantitative lifetimes and pure dephasing times of the image states at different states in k-space were obtained by fitting the correlation data with solutions for the three-level optical Bloch equations. A model explaining the k-dependence lifetime of image states is presented. This work was supported by the Army Research Office and the Columbia JSEP Program.

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Abstract Submitted
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Two-Photon Photoemission Study of Stepped Cu(111) Surface Using Femtosecond Laser Pulses X. J. SHEN, HIDONG KWAK, A. M. RADOJEVIC, R. M. OSGOOD JR., Columbia Radiation Laboratory, Columbia University — Multiphoton photoemission using ultrafast laser pulses is a powerful technique for probing carrier dynamics at surfaces and interfaces. Angle- and time-resolved two-photon photoemission measurements were performed on stepped single-crystal copper surfaces with features of ~14 Å terrace width and ~2 Å step height. The experimental setup used a variable time delay between a femtosecond pump (~267 nm and 100 fsec) and probe (~400 nm and 80 fsec) pulse, which were generated by frequency doubling and tripling the laser pulses from a Ti:sapphire fsec laser system. Based on the optical Bloch equations for a three level system calculations, we were able to obtain the lifetimes of the n=1 image state electrons by fitting the correlation data. Electrons with bigger momentum parallel to the surface had a shorter lifetime, which we attributed to the total effects of the presence of the step potentials and the energy resonance with empty bulk states. A comparison of the electronic relaxation dynamics between flat and stepped Cu(111) surfaces will be discussed. Financial support of this work by the Army Research Office and the Columbia JSEP Program is gratefully acknowledged.

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